Story of Ruby Maser (Commemoration Issue Dedicated to Professor Sakae Shimizu on the Occasion of his Retirement)

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Note

Story of Ruby Maser

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A brief personal account of the Ruby Maser Research and Development Program is given, to point out the time and the web of technical environment that led to its development at Willow Run Laboratories.

KEY WORDS Ruby / Maser /

In the Smithsonian Institution, Washington, D. C., there now hangs the frame containing the "World's First Maser Ruby", with the inscription

Maser Action Was First Produced
In This Linde Synthetic Ruby
On the 20 December 1957
At Willow Run Laboratories.
Sample Donated by Mineralogy Department
Of the University of Michigan

This event, which later led to the development of ruby masers for a number of astronomical applications, such as for the radiometry of planetary microwave radiations and the reception of close-up pictures of Mars, took place in the rear of a converted hangar, Bldg. 41, near the northest end of the runway of the Willow Run Airport.[1,2]

On this occasion of commemorating the retirement of Professor Sakae Shimizu, in whose laboratory I spent a very pleasant 1969-70 academic year, it occurred to me that it might be appropriate to review some of the events surrounding the research and development programs of the maser research and development programs at the Willow Run Laboratories, to see what lessons we might be able to learn from the past events. In the brief span of two decades, the peak of the golden era of U. S. science research has passed; instead Japanese science research seems to be on the ascent.

In 1959, when I first visited some of the laboratories in Japan, I was appalled by the conditions under which some researchers were struggling. The one that I still remember vividly was a visit to Hiratsuka, I believe, where a Cobalt-60 source was kept in a well in the middle of a rice paddy and certain semiconductor measurements were being carried out in what looked like an abandoned factory. Things are now very different, and my hope is that the comments to follow will help those who will be

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leading Japanese science during the decade or two to come.

The maser research and development program at the Willow Run Laboratories owes its success to the unusual combination of administrative and technical conditions created by the atmosphere for research during the decades of 1950's and 60's. The program was initiated at the suggestion of Weston E. Vivian, who later served in the 89th Congress (1965-66), and approved by Newbern Smith, the then Chief Engineer of the Willow Run Laboratory, and by Lloyd Mundie, the head of the Infrared Group. The discussions took just a few days and was approved initially at the level of $60,000 for the first year.

In the fall of 1955, the Solid State Group, in which the maser program was to be carried out, was just getting started under the direction of Dr. Lyle Tiffany, so that there was then essentially no research equipment and of course, essentially no technical know-how. For example, the use of liquid nitrogen, which was being done routinely in other U. S. research laboratories, was still a few months away. And, of course, the Varian EPR spectrometer was still not on the market, so that our first order of business was to put together an EPR (or ESR, for Electron Spin Resonance) spectrometer by buying an electromagnet and a variety of microwave and electronic components. It was clear from the beginning that we would need to build equipment for experiments at liquid helium temperatures, but such concerns had to be deferred in face of the more urgent immediate needs. All of this, of course, meant time and money. Just a month after the project got under way, I remember receiving a phone call from the front office notifying me that I had spent already $20,000 and was reminded that the budget limit was $60,000. That, however, was the last phone call, despite the fact that by the end of the year, I had overspent by a factor of two, if not three.

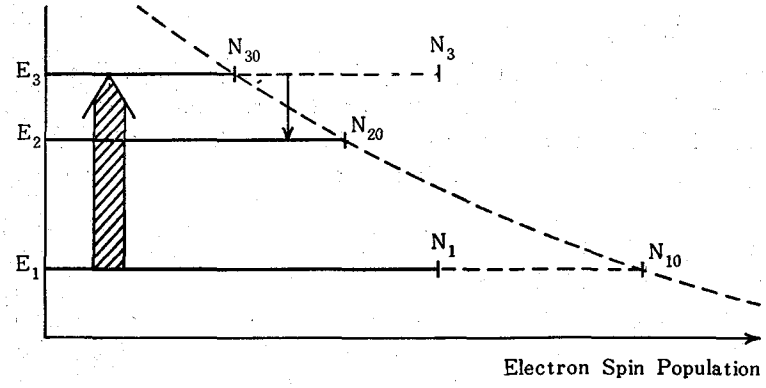
But possibly the more critical problem was lack of skilled technicians. I was authorized to hire a technician, and after many interviews, I chose Dick Ager, who then had just finished a radio repair school and whose only previous experience was working in his father's candy store.

It should be remarked that these conditions that existed in and around the Willow Run Laboratories were quite different from those in other research laboratories on the Eastern Seaboard of the United States, such as those of Bell Telephone, Lincoln, Columbia, and Harvard, where there were aleady well-established research groups in electron spin resonance. The "edge" that the research workers in those laboratories had over us can be inferred from the fact that Bell Telephone workers, H. E. D. Scovil and his co-workers,[3] reported the results of their sophisticated experiment on gadolinium ethyl sufate in November of 1956, when we were still putting together our ESR spectrometer. About half a year later, in July 1957, A. L. McWhorter and J. W. Meyer[4] of the Lincoln Laboratory, reported their success with chrome cobalti-cyanide. At Willow Run, then, we were still designing and waiting for cryogenic equipment for liquid helium. The first liquid helium transfer came about four months later, in November of 1957.

Although the development of the experimental facilities was coming along at an agonizingly slow pace, the calculations and the design of the ruby maser experiment was completed in a matter of few days near the beginning of 1957.

The word MASER (not to be confused with LASER) is an acronym for Microwave Amplification by Stimulated Emission of Radiation), coined by C. H. Townes for his

ammonia beam device.[5] A short time later, N. Bloembergen[6] foresaw the possibility of designing a microwave amplifier by means of a paramagnetic solid. The proposed idea can perhaps be best illustrated by means of the following diagram:



In the above diagram, E_1, E_2, and E_3 are three of the $2S+1$ ground state Zeeman levels of a paramagnetic ion of electron spin S, and N_{10}, N_{20}, and N_{30} are the corresponding electron spin populations. If a signal frequency at $hf=E_3-E_2$ is applied, near equilibrium N_{20} is greater than N_{30} so that the paramagnetic material will absorb energy at the microwave frequency. But if by some means or another the population of level 3 could be made to exceed that of level 2, then the applied signal would be amplified. Bloembergen's suggestion was to bring about such an electron spin population inversion by "pumping" electron spins from a lower level, such as level 1, increasing the population of level 3 to N_3.

But to effect this pumping, it is necessary to break down the selection rule so that transitions level 1 to level 3 can occur. To see how this can be accomplished, consider the spin Hamiltonian

$$H=g\beta S\cdot H+D\left\{S_z^2-\frac{1}{3}s(s+1)\right\}$$

which is applicable to high degree of accuracy to many paramagnetic ions. The first term is the Zeeman coming from the interation of the electron spin with the applied magnetic field the second can be thought of as an effective crystalline quadrupolar field. It now D were zero, it can be easily shown that the Hamiltonian will commute with the sp in operator S_z ; and for such conditions, only transitions between adjacent Zeeman levels will be allowed. On the other hand, if $D\neq 0$,

$$[H,\ S_z]=\frac{g\beta}{2}[H_+S_-+H_-S_+,\ S_z]$$

$$=\frac{g\beta}{2}(H_+S_--H_-S_+)\neq 0$$

A very special case occurs when the applied magnetic field is parallel to the crystal axis. Then both H_x and H_y are zero, so that then the eigenfunctions of S_z will also be eigenfunctions of the Hamiltonian. In the realm of "basic research" this condition is used for the measurement of the parameters such as g and D, because then only tran-

sitions between adjacent magnetic states will be allowed, the absorption spectrum will be the simplest, and therefore the easiest to interpret.

If, however, the magnetic field is at an angle with the magnetic field, although S_z will not commute with the Hamiltonian, but the commutator

$$[H, S^2]=0$$

so that now the eigenfunctions of the Hamiltonian will be given by a linear combination of the $(2S+1)-$fold degenerate eigenfunctions of the spin operator S^2. This admixing of the spin functions leads to the break down of the selection rule $\Delta M=\pm 1$ and to the possibility of pumping. Furthermore, it is clear that pumping is easier the larger the value of D.

For a maser material, there are two contradictory requirements, namely the high concentration of paramagnetic ions needed for high amplification, but a low concentration to assure sufficiently long spin-lattice relaxation time. Since at liquid helium temperatures all spins will be in the ground multiplet, and all levels of this multiplet will be essentially equally populated even at such low temperatures, the population of any level will be given by $N_c/(2S+1)(2I+1)$, in which N_c is the concentration of the paramagnetic ion in the solid, S is the electron spin as mentioned earlier, and I is the nuclear spin of the paramagnetic ion. Since the nuclear spin plays no part in the maser, ideally the nuclear spin should be zero. Also S needs to be as small as possible, but consistent with the fact that we need to have at least three levels. This means then that S needs to be at least 1 or possibly 3/2. Then the candidate ions in the iron group are Ni^{++} $(S=1)$ and $Cr^{+++}(S=3/2)$. There is another well known ion $V^{++}(S=3/2)$, but vanadium consists essentially 100% of V-51, whose nuclear spin is 7/2. For chromium, in contrast the bulk of the isotopes are even with zero nuclear spin, with only 9.5% of Cr-53 $(I=3/2)$.

Consequently, in our search for a good maser material, attention was focused on paramagnetic materials containing nickel or chromium, with large zero-field splitting, D. Furthermore, in addition to these "theoretical" requirements, there was the "practical" requirement that the maser material would need to be resistant to thermal shock. Before 1957, the bulk of the research on paramagnetic materials had been on hydrated salts, grown from aqueous solutions and which inevitably contained pockets of water. This could cause shattering upon repeated cooling and warming. Consequently it was felt that the material suitable for a practical maser would need to be some "hard" crystal with good thermal shock properties, containing no water, possibly not even hydrogen. Fortunately, it happened that I had spent two years at the U. S. Naval Research Laboratory, where Dr. L. Matarrese and I had started some measurements on ZnS : Mn and had begun to see the potentialities of using ESR for the study of such non-hydrated cyrstals.[7] Also in the same laboratory, L. Singer was making measurements on chromium trichelate and more than once I heard him comment about the large D value.[8] It was about this time, while pondering about the courses of action to take, that the paper by J. E. Geusic on "Paramagnetic Fine Structure Spectrum of Cr^{+++} in Single Ruby Crystal" came to my attention.[9] Upon further study of this material, I learned about the exceptional thermal conductivity properties of sapphire (α-Al_2O_3) at liquid helium temperatures, so the decision was made to use this material for our first attempt to see maser

action.

As mentioned earlier, the design for the experiment took only a few days. The spin-Hamiltonian given above leads to the eigenvalue equation

$$(\varepsilon-1)^2-\frac{5}{2}x^2\varepsilon^2-2\varepsilon x^2(1-3\cos^2\theta)+\frac{9}{16}x^4-x^2(6\cos^2\theta-1)=0$$

$$\varepsilon\equiv\frac{E}{D},\qquad x\equiv\frac{g\beta H}{D}$$

which is quartic in the eigenvalue ε. At that time at Willow Run, computing facilities had not yet been fully developed, I personally did not want to become involved with numerical solutions, what would have been necessary for small angles around 15°, but noticed that the quartic equation could be reduced to a bi-quadratic one for

$$3\cos^2\theta-1=0$$

or near 55°. The solutions for this case are

$$\varepsilon\left(\pm\frac{3}{2}\right)=\pm\sqrt{1+\frac{5}{4}x^2+x\sqrt{3+x^2}}$$

$$\varepsilon\left(\pm\frac{1}{2}\right)=\pm\sqrt{1+\frac{5}{4}x^2-x\sqrt{3+x^2}}$$

These then led to the following comments that appeared in the Infrared Group Progress Report for January 1957:

SPIN RESONANCE AMPLIFIERS : Probl. 43
MASER

Some of the calculations necessary for the MASER (Microwave Amplification by Stimulated Emission of Radiation) cavity design have been carried out. Because the critical frequency of ruby is 11.4 KMC/sec, it was decided to design a cavity appropriate for Bloembergen's three-level MASER system. Our calculations indicate the following results:

Signal to be amplified = 9.3 KMC/sec
Energizing frequency = 24 KMC/sec
Magnetic field = 38 qauss*
Cavity diameter = 1.86"
Signal frequency mode, circular, TE_{011}
Energizing frequency mode, circular TE_{114}

*This no doubt was a typing mistake. The value was most probably 3,800 gauss.

It appears that we had begun to test our dual mode cavity around March or April. For it happened that I was invited by Gerhard Weibel in April 1957 to discuss our

maser research and development program at the Sylvania Research Laboratory, then in Bay Shore, Long Island. The topic I chose was "MASARS and MASERS". When I met him again some months later, he thanked me for having disclosed completely the plans I had mapped out.

A crucial turn in our program took place when one day, both George Makhov, then a graduate student in electrical engineering, and I went to the Mineralogy Department with the hope of obtaining a sample of ruby. As Professor R. M. Denning scrounged through his drawers for pieces of ruby, he casually asked whether synthetic ruby would do. It happened that he had been making measurements on the elastic properties of Linde ruby. The difference, he pointed out, was that the color is substantially lighter, due to the much smaller concentration of chromium. The implications were so obvious that we took as much as he could spare.

As mentioned earlier, we read in August about the success at the Lincoln Laboratory, we carried out our first liquid helium tansfer in November, and after the usual debugging period, we observed maser action in the pink ruby about a month later, on December 20.[10]

Since that date, much of what others had done in the laboratory in the rear of Bldg. 41 have slipped from my memory, except G. Makhov's (now deceased) attention to details, J. Lambe's (now Ford Scientific Laboratory) uncanny ability to make experiments work, and R. W. Terhune's (also Ford) superb ability to develop needed instrumentation.

Almost immediately, the decision was made to explore the capabilities of the ruby maser for the measurement of astronomical microwave sources. This part of the program was continued under the supervision of Dr. R. W. Terhune. Two years later Bair, Cook, Cross, and Arnold, working also at the Willow Run Laboratories,[11,12] reported the striking results on microwave radiations at 3.45 cm from Cassiopeia A and Tycho Brahe's Supervova.

The developments that have attracted much public attention are the closeup pictures of planets such as Mars, made possible by the ruby maser built by Walter Higa and his co-workers of the Jet Propulsion Laboratory.[13] Some of the initial results were obtained from the flight of Mariner 4 in 1965 but the more spectacular photographs were by Mariners 6 and 7 in 1969,[14] about 14 years after Bloembergen first suggested the solid-state maser and about 12 years after the success at Willow Run.

The point is that technological progress takes time. Historians will say that "Rome was not built in a day"; yet the same historians expect the miracles of technological breakthroughs. The American public, led by the press into the belief of technological breakthroughs has unfortunately become impatient with progress made in the name of scientific research. Much of the fault is with the U. S. mass media; but a part of the fault is ours for having not stressed the time it takes to accomplish our achievement. My hope is that our colleagues in Japan will not make the same mistake. By stressing the time it takes to make scientific progress, perhaps we can educate the public, through the press, that scientific progress, like political developments, come about very slowly.

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